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**FILTER MATERIAL EFFICIENCY
AGAINST HIGH LEVELS OF AIRBORNE
RADIOACTIVE CONTAMINATION**

by

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Jerry J. Mahoney
Robert B. Price

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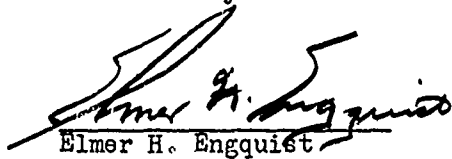
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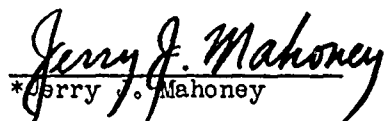
FILTER MATERIAL EFFICIENCY AGAINST HIGH LEVELS
OF AIRBORNE RADIOACTIVE CONTAMINATION

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Notebooks: 3055, 3092, 3180

Experimental work:
Started: 19 July 1949
Completed: 13 March 1951


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Typed: 13 August 1951
ec, pmc

Control no.
5022-14

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ABSTRACT

Object.

The object of project 4-12-75-001 is to evaluate the problem of providing adequate protection against radiological hazards. This includes the evaluation of current military protective measures and equipment developed for BW and CW protection and determining the limitations, if any, in their use for RW protection.

The object of the work described in this report was to determine the efficiency of Chemical Corps type 6 filter material against airborne radioactive particulates of a radioactivity level of the order of magnitude of the activity anticipated in the cloud in the vicinity of an atomic-bomb detonation.

Results.

The efficiency of Chemical Corps type 6 filter material with levels of activity from 3.0 to 4,000 μ c./l. was evaluated using the test method, with some modifications, reported in TCR 60 (18 April 1950). The method involves the use of sodium iodide particulates labeled with I^{131} ; the particulates of the test medium have a mean particle size of 0.48 μ .

The filter material permitted the penetration of only 0.020% to 0.045% of the influent test agent up to activity levels of 20 μ c./l. At activity levels of 60 μ c./l. and 4,000 μ c./l., the efficiency increased to permit the penetration of 0.005% to 0.007%. Accuracy of the above measurements by radiometric methods was within $\pm 20\%$.

Conclusions.

1. The Chemical Corps type 6 filter material may be effectively used against airborne radioactive particulates of a radioactivity level of the order of magnitude of the activity anticipated in a cloud in the vicinity of a nominal atomic detonation at H + 10 min.

a. The penetration of type 6 filter material by an aerosol with a 0.6 Mev beta radiation and a 0.37 Mev gamma radiation in activity levels up to 20 μ c./l. is approximately 0.02% to 0.04%.

b. Between 60 μ c./l. and 4,000 μ c./l. aerosol activity, the penetration efficiency is approximately 0.005% to 0.007%.

2. There is a significant increase in efficiency of type 6 filter material for the removal of an aerosol in activity levels of 60 μ c./l. and 4,000 μ c./l.

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Recommendations.

None, since work is continuing under this project.

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FILTER MATERIAL EFFICIENCY AGAINST HIGH LEVELS
OF AIRBORNE RADIOACTIVE CONTAMINATION

I. INTRODUCTION.

A. Object.

The object of project 4-12-75-001 is to evaluate the problem of providing adequate protection against radiological hazards. This includes the evaluation of current military protective measures and equipment developed for BW and CW protection and determining the limitations, if any, in their use for RW protection.

The object of the work described in this report was to determine the efficiency of Chemical Corps type 6 filter material against airborne radioactive particulates of a radioactivity level of the order of magnitude of the activity anticipated in the cloud in the vicinity of an atomic-bomb detonation.

B. Authority.

Authority for this work was contained in the project programs for fiscal years 1950, Project 4-12-01-02, Evaluation of Protection for RW, and 1951, Project 4-12-01-002, Evaluation of Protective Equipment and Materials Against RW Hazards. The number of this project was changed to 4-12-75-001, CCTC item 2214, 20 Nov. 1950; the title was changed to Protection Against Radioactive Gases and Aerosols, CCTC item 2324, 25 May 1951.

II. HISTORICAL.

The background information on filter testing methods and apparatus and the penetration efficiency for filter material for aerosols up to 20 μ c./l. is summarized in a previous report (1). Studies of effects of radioactivity on penetration are available (2), but deal mostly with tracer levels of radioactive aerosol.

III. THEORETICAL.

The several mechanisms involved in the filtration process are described in a previous paper (1), with some consideration of possible effects of high levels of radioactivity in aerosols on these mechanisms.

Effects of electrostatic charges and ionization on filtration may be appreciable, especially at high radioactive levels. Victor K. La Mer stated in a recent seminar (May 1951) at Army Chemical Center that ~~these~~ electrostatic effects are being investigated.

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IV. EXPERIMENTAL.

A. Materials and Equipment.

1. Isotope.

The isotope used for this test was I^{131} , which has a half life of 8.0 da. The following radiations are present in the decay: 0.6 Mev beta radiation, 0.367 Mev and 0.080 Mev gamma radiation. This isotope was obtained from the Isotopes Division AEC, Oak Ridge. (Catalog No. 3 July 1949, Item No. S-2).

2. Test Apparatus.

a. Atomizing Equipment.

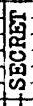
A schematic diagram of the test apparatus is shown in fig. 1, and the photographs of the principal items, unassembled, of the test apparatus in figs. 2 and 3. Except where modifications are specifically mentioned, the apparatus and procedure are the same as those previously described⁽¹⁾. The test apparatus is shown unassembled because of the great number of lead bricks used for shielding when assembled.

Several modifications were made in the basic design to meet the high-activity requirements of 4 mc./l. of aerosol. A new stainless-steel atomizer of the same dimensions as previously described was made but with only one atomizing hole instead of the standard three holes. A stainless-steel hypodermic needle was made to extend the one-hole intake at the bottom of the atomizer. This was necessary to draw the liquid from the small calibrated glass well at the bottom of the atomizing solution bottle. Usually exactly 4.0 ml. of atomizing solution was used at the start of a run.

Type 6 filter material was utilized also for collecting influent and effluent samples for counting purposes. It is presumed in calculations that the No. 6 effluent samples collected 100% efficiently. Since ratios of effluent to influent samples were used, the error introduced by this assumption is negligible. The diameter of the sample sheets was 1 in., two such always being used in parallel. To maintain the standard linear flow rate of 320 cm./min. through the 1-in. diameter filter, 1.6 l./min. was required. The aerosol generated was then always 3.2 l./min. No additional mixing air was used. It was found that 34 lb./sq.in. pressure was required to give this volume of air through the atomizer. The atomizer used 0.1 ml./min. of solution at 34 lb./sq.in. pressure.

A remote pipetting device (Tracerlab Model E-18A) was used to transfer the atomizing solution. This pipetting device was modified by cutting off its handle and turning through 90°. This allowed operation behind a 3-in. thick lead brick wall by using mirrors. The modified pipette holder is shown in fig. 2. The hands or eyes of the operator were thus never in the line of sight of the radiation.

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FIGURE 2

TEST APPARATUS

Top Photograph: Remote pipetting device and atomizer.

Bottom Photograph: Muffle furnace and drying tube for aerosol.

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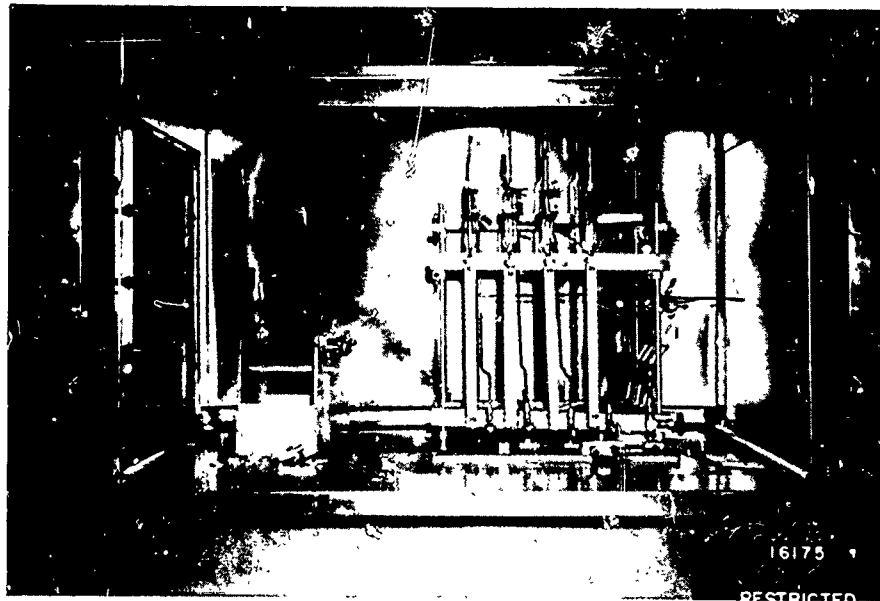
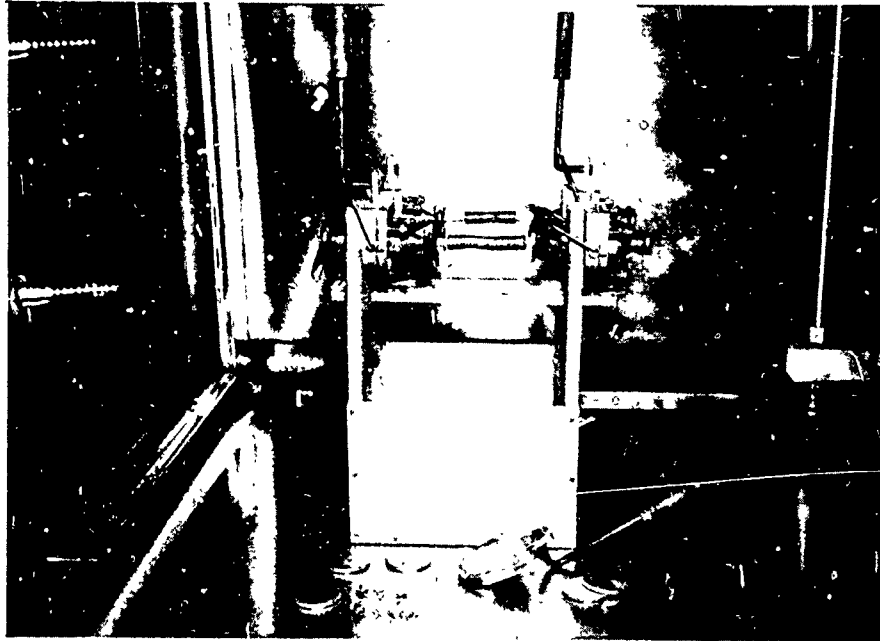


FIGURE 3

TEST APPARATUS (Continued)

Top Photograph: Filter sample holders.

Bottom Photograph: Flowmeter system for measuring aerosol volume.

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b. Distillation Equipment for Atomizer Solution Concentration.

It was necessary to concentrate the atomizing solution to obtain the high radioactive aerosol levels required. Distillation was considered the best method.

Preliminary tests were made to determine if any I^{131} was lost when the atomizer solution was concentrated by boiling down. For such a test a 2% solution of inert sodium iodide in water was made. To 4 ml. of this solution, any required amount of I^{131} sodium iodide solution was added. The Oak Ridge figure for this activity was used. The Oak Ridge figures for activities of shipments have been spot checked repeatedly (1). Using infrared lamps, enough of the solution was distilled off to leave exactly 4 ml. of solution in the atomizer bottle. A visible amount of solid was deposited on the inside walls of the bottle during process. This was anticipated but had a negligible effect on the volume of the liquid left after boiling. This surface deposit dissolved almost instantly upon commencing the atomization. Numerous tests showed that this method of volume reduction did not cause a measureable amount of I^{131} to be lost. This was determined by testing filter samplers, either effluent or influent, after boiling down several different concentrations of NaI solution, I^{131} labeled, and counting samples. The results followed the simple relationship of direct proportion of counts/sec. in the airborne collections to solution radioactivity. A sample calculation is shown in appendix A.

In addition to distillation of initial solutions, the solution concentration increased some with time during the atomization. Using collected samples on filter material as a measure of this increase in concentration, tests at several radioactive levels were made. The per cent change in concentration during the atomization was quite consistent over levels giving a range used in this report of 10 to 200 counts/sec. on sampling filters as shown in figs. 4 and 5. These examples were made using the standard procedure for 4 ml. of initial volume of atomizing solution. An initial volume of 4 ml. was used for all final results tabulated in this report. This increase in solution concentration with time, although about 20%, was considered inconsequential as final penetration efficiencies were calculated from ratios of influent and effluent samples taken simultaneously.

c. Counting Apparatus.

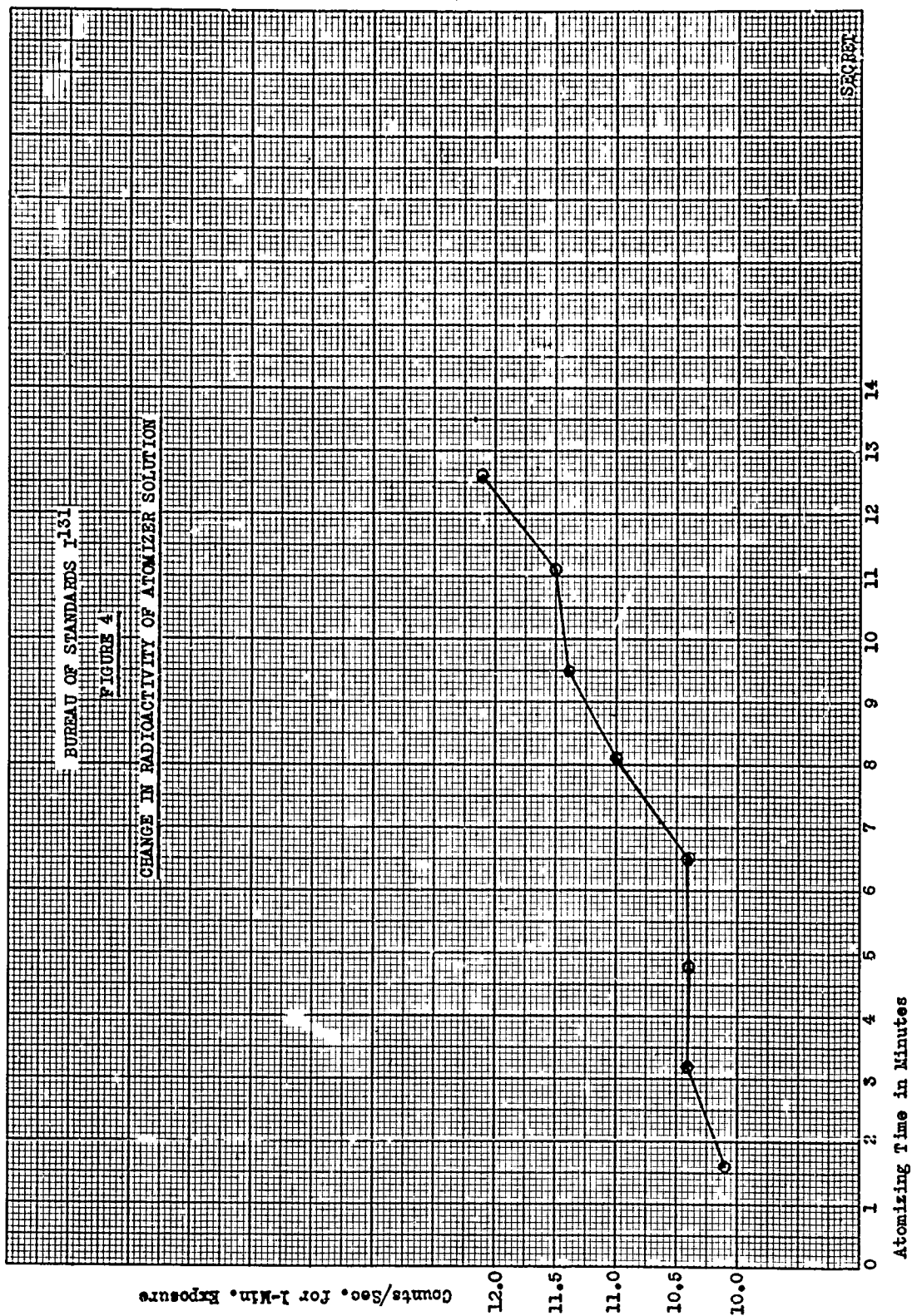
A Tracerlab Autoscaler Model SC-1 was used in counting the filter-material samples. The Geiger-Mueller (G-M) tube used was a RCL Mark 1, Model 3, having a window thickness of 2.8 mg./sq.cm.

B. Procedure.

1. Operation of the Apparatus.

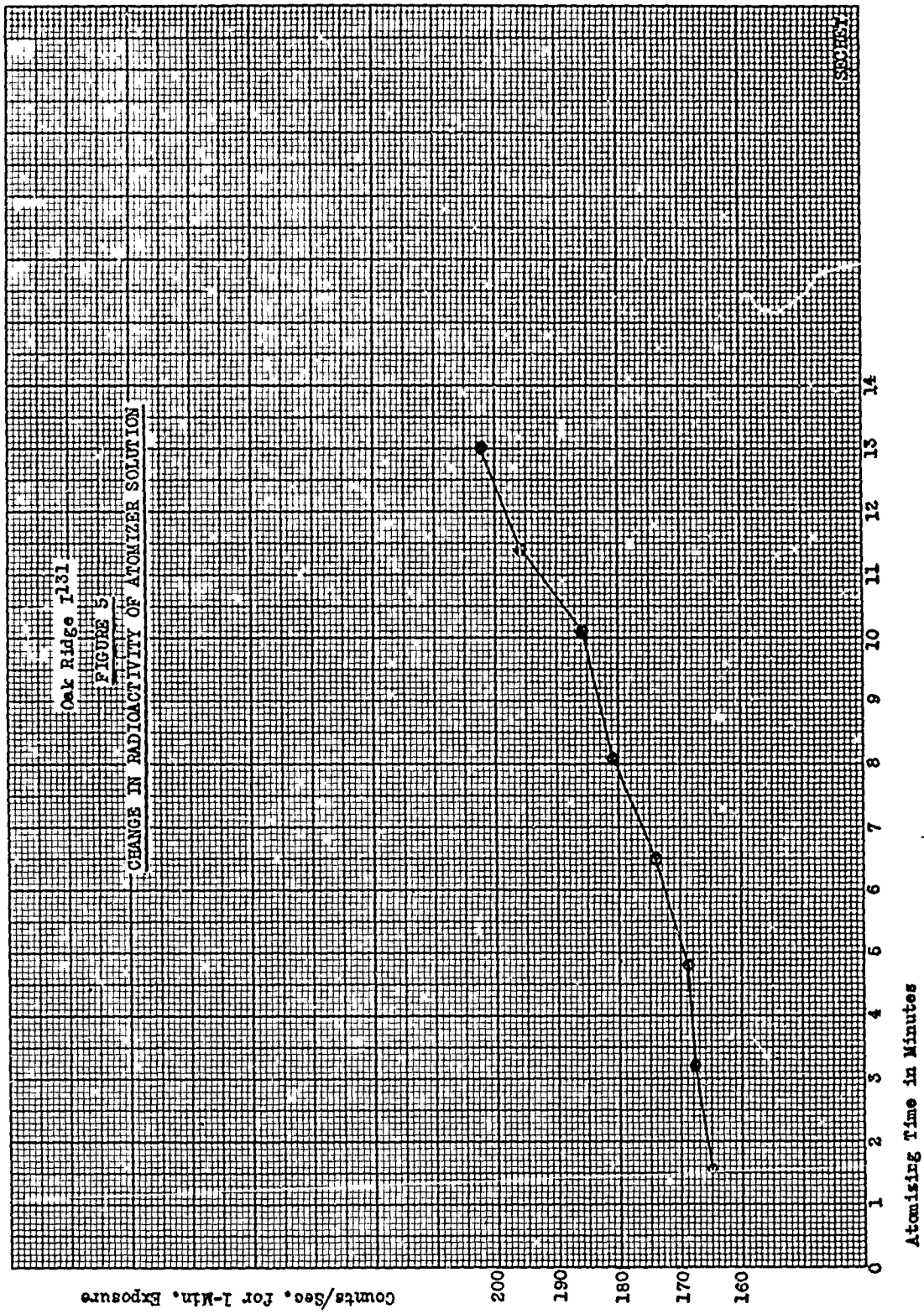
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STEP 1

A measured volume of the Oak Ridge NaI¹³¹ solution was pipetted into the atomizer bottle through a separatory funnel. The volume used was determined by the number of millicuries to be used in any particular run.

STEP 2

Four milliliters of inert 2% NaI was added through the same separatory funnel.

STEP 3

Approximately 10 ml. of distilled water was added for additional rinsing of the separatory funnel. The stopcock on the separatory funnel was opened and closed by a brass rod through a hole in the 3-in. thick lead brick wall lining the operating laboratory hoods.

STEP 4

The separatory-funnel stopcock was closed and left closed for the following operation. The two heat lamps focused on the glass atomizer bottle were turned on, and at the same time a vacuum "pull" was started through the auxiliary "by-pass", shown in fig. 1. The material in the atomizer bottle was boiled down to a volume of 4 ml. indicated visually with mirrors showing the markers on the calibrated well at the bottom tip of the atomizer bottle. The compressed air supply to the atomizer was left off during the boiling-down process.

STEP 5

The infrared lamp and the auxiliary by-pass were shut off and left off for the succeeding run. The atomizer was allowed to cool to room temperature.

STEP 6

While the 4 ml. in the atomizer were cooling, the muffle furnace surrounding the drying tube was turned on. A preset rheostat in the heating circuit was used such that the thermometer at the exit end of the drying tube registered 130°C. The air entering at stopcock "A" was "pulled" by vacuum through the equipment and "dummy" filter papers in the samplers to bring the apparatus to steady conditions. About 1 hr. was adequate for this process, the atomizer bottle being cooled to room temperature in the same time.

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STEP 7

The air flow was switched from the filter samples to the regular by-pass.

STEP 8

The "dummy" filters were removed and discarded. Fresh filters to be tested were placed in all four holders.

STEP 9

The compressed air for atomizing was turned on and the aerosol allowed to flow for about 1 min. through the main by-pass.

STEP 10

The gang stopcock was turned through 90°, instantly shutting off the by-pass and putting the aerosol through the sample filters to be tested. The samples were all subjected to 1-min. exposures except the filter paper in holder No. 4, fig. 1. This No. 4 holder was left on continuously as it held the filter under test for efficiency against the total activity.

STEP 11

At the end of exactly 1 min. of collection, the gang stopcock was switched to by-pass. The "hot" filters were immediately removed with tweezers and new filters inserted. This was done as rapidly as possible as only 18 min. of atomizing time was possible with the 4 ml. used. During atomizing about 2 ml. of atomizer solution was suspended in air or flowing down the inside of the atomizer such that after about 18 min. of operation the tip of the hypodermic needle was not covered by liquid. Ten good 1-min. samples could be obtained during the 18 min. of operating time.

STEP 12

The effluent filters were placed directly in aluminum cups with friction lids for subsequent counting. The influent samples were placed in 50 ml. of distilled water in beakers for dilution and counting as described in section 3. For the high-level runs the filters were too radioactive for direct counting. Any dilution thereof is shown in the data. In the data all even-numbered filters such as 2E, 6E, etc. were from the same branch as the No. 4 filter holder. All odd-number filters are from the arm containing holders Nos. 1 and 3. In the odd-numbered arm the influent sample in holder No. 3 was changed for each minute of exposure. In the notation, "E" stands for effluent and "I" for influent. As an example, 4E means effluent sample No. 4, while 7I means influent sample paper No. 7, and so on.

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2. Method of Counting High-Activity Influent Filter Samples.

STEP 1

Filter samples on the two influent sample collectors were taken simultaneously to give about 400 counts/sec. when the aluminum cup containing the sample was on the lowest counting shelf.

STEP 2

One sample was then counted directly in the usual way.

STEP 3

The second sample of filter paper was placed in 50 ml. of distilled water to dissolve the NaI.

STEP 4

One-milliliter samples of the step 3 liquid were pipetted into open Tracerlab pyrex counting cups 1 in. in diam. and 1/4 in. deep.

STEP 5

These 1-ml. samples of step 4 were evaporated by heat lamps, allowed to cool, and counted.

STEP 6

As the result of numerous tests and interchanging the arms of the collecting system, an average counting ratio of 37.5 was found. This means that "hot" filter papers could be placed in 50 ml. of distilled water, sampled as above, and counted. This count multiplied by 37.5 gives what this "hot" filter would count in the standard aluminum cup. A sample determination of this ratio is shown in appendix B.

A few remarks may be useful in clarifying some points in the above dilution ratio determination. At 400 counts/sec. the resolving-time correction of the G-M tube is not in excess of 8% for a resolving time of 2 μ s. The effect of the 0.006-inch-thick aluminum counting cups need not be considered, as absolute counts/sec. or absolute millicurie value of the sampling filters was not required. Only ratios of influent to effluent counts/sec. under the same collecting conditions were used in calculating the final penetration efficiencies. Further, in all cases effluent and influent samples were counted within an hour or two of each other so the "decay" factor for I¹³¹ was negligible in this time interval.

Extension of the dilution method was tested. It was found that dilution by still another factor of 100 was possible to

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the same degree of accuracy, $\pm 3\%$ in the ratio 37.5. This greatest dilution makes a conversion factor of 3,750 necessary for the greatest experimental radioactivity used; namely about 4 mc./l. of aerosol gas. There were five shelves in the lead "pig" used to hold samples while counting. Number 5 was the bottom shelf farthest from the G-M tube. The 400 counts/sec. mentioned above were counted on the lowest shelf. The geometry ratio in counting for the shelves in the "pig" was taken frequently, and always when any change in the counting apparatus was made such as the installation of a new G-M tube. A sample geometry determination is shown in appendix C.

3. Analysis of Filter Material.

A set of data covering two different 1-min. exposures of test filters is shown in the table below.

Sample Calculation of Penetration

Sample number	Total counts for 1 min. of exposure	Total time sec.	Counts/sec.	Counts/sec. corrected for back-ground	Counting shelf number	Counts/sec. converted to regular aluminum cups on shelf 5	Penetration
5I	12,288	99.8	123.1	122	5 in glass cup	45,750	0.0077
6I	Not counted	-	-	-	None	45,750 assumed	0.0077
5E	384	62.2	6.17	5.57	4 in Al cup	3.53	0.0077
6E	384	62.5	6.14	5.54	4 in Al cup	3.51	0.0077
13I	12,288	94.5	130.0	129	5 in glass cup	48,375	0.0052
14I	Not counted	-	-	-	None	48,375 assumed	0.0068
13E	384	83.6	4.59	3.99	4 in Al cup	2.53	0.0052
14E	384	66.1	5.81	5.21	4 in Al cup	3.30	0.0068

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C. Results.

The results were calculated on the basis of efficiency of penetration as given by the following equation:

$$\% \text{ Penetration} = \frac{\text{Counts/sec./l. (effluent)}}{\text{Counts/sec./l. (influent)}} \times 100$$

The results are given in the chronological order of tests.

1. Activity Level - 20 μ c./l.

The efficiency for the two samples tested was 0.020% and 0.019%. This was a "pilot" run.

2. Activity Level - 60 μ c./l.

These data are shown in fig. 6. The average penetration is about 0.007%.

3. Activity Level - 250 μ c./l.

These data are shown in fig. 7. The average penetration is about 0.006%.

4. Activity Level - 4 μ c./l.

These data are shown in fig. 8. The average penetration is about 0.02%.

5. Activity Level - 250 μ c./l.

These data are shown in fig. 9. The average penetration is about 0.007%.

6. Activity Level - 4,000 μ c./l.

This is the final high-level test. A total of 1.12 curies was used. The results are shown in fig. 10 and give a penetration of about 0.005%.

7. Activity Level - 3 μ c./l.

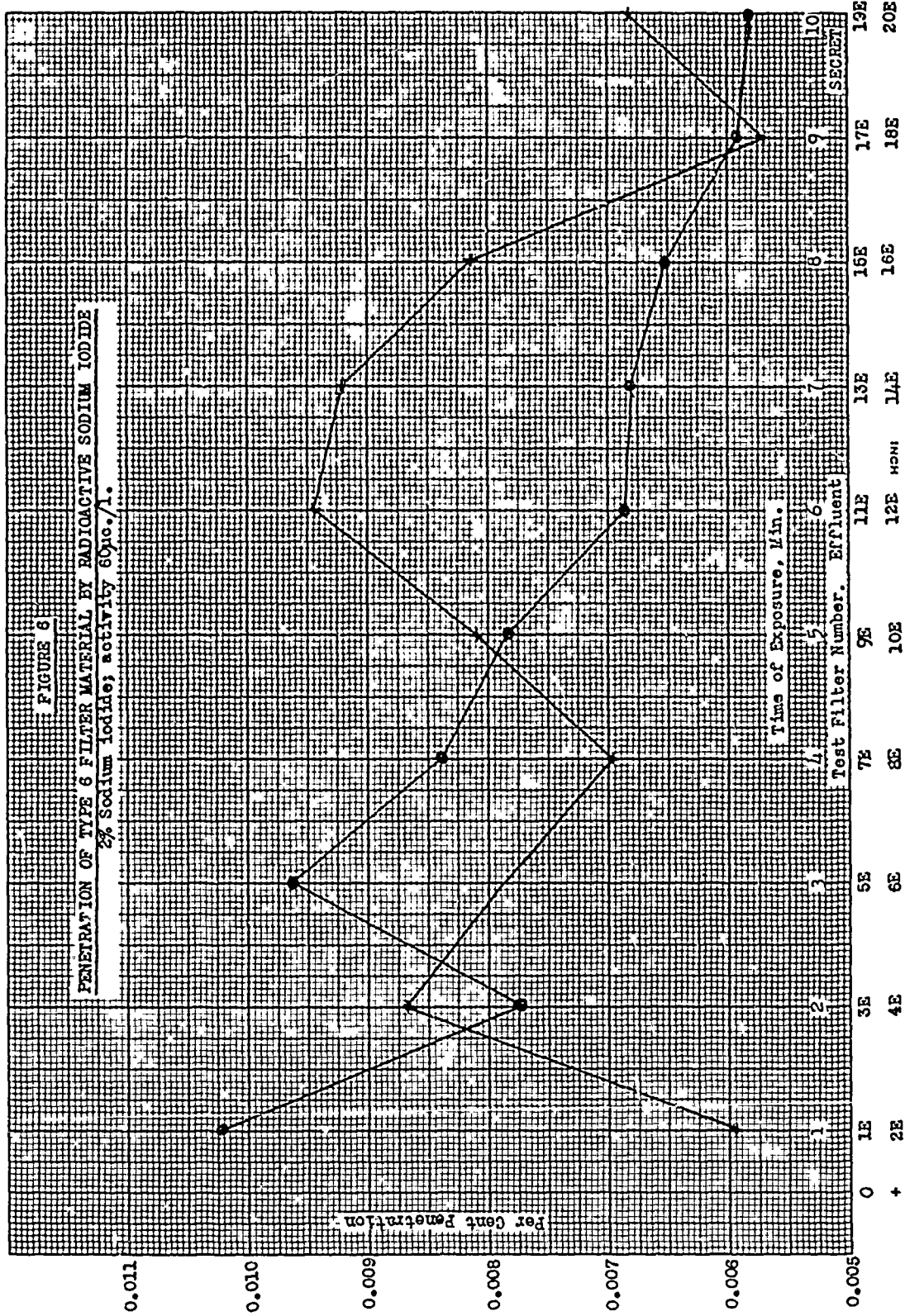
After decontamination of equipment, this low level was used as a recheck. The results are shown in fig. 11, giving an average penetration efficiency of 0.03%, the normal result.

8. Saturation Effect of Test Filter.

No observable effect of plugging of the tested filter in holder No. 4 of fig. 1 occurred. The penetration efficiencies for

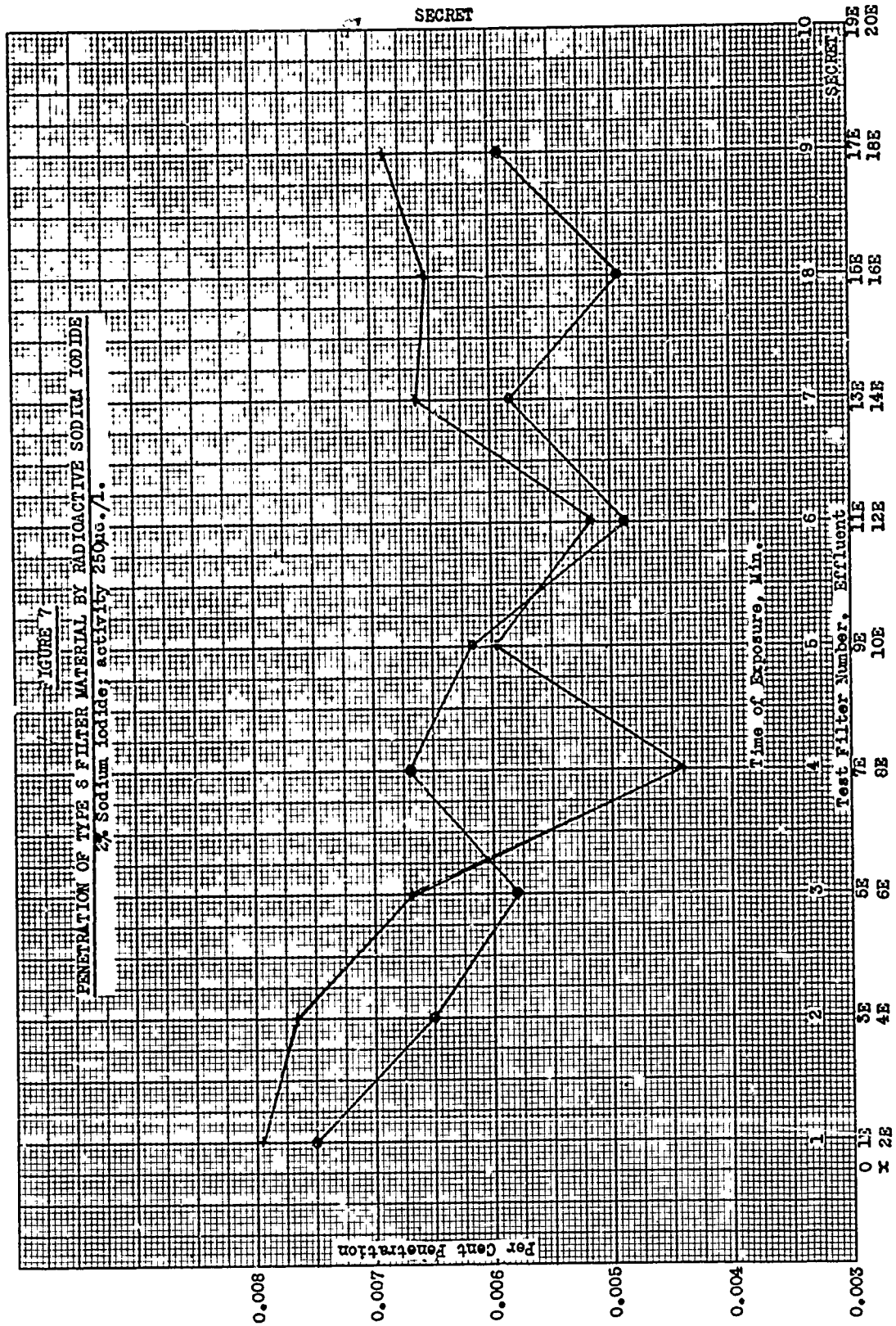
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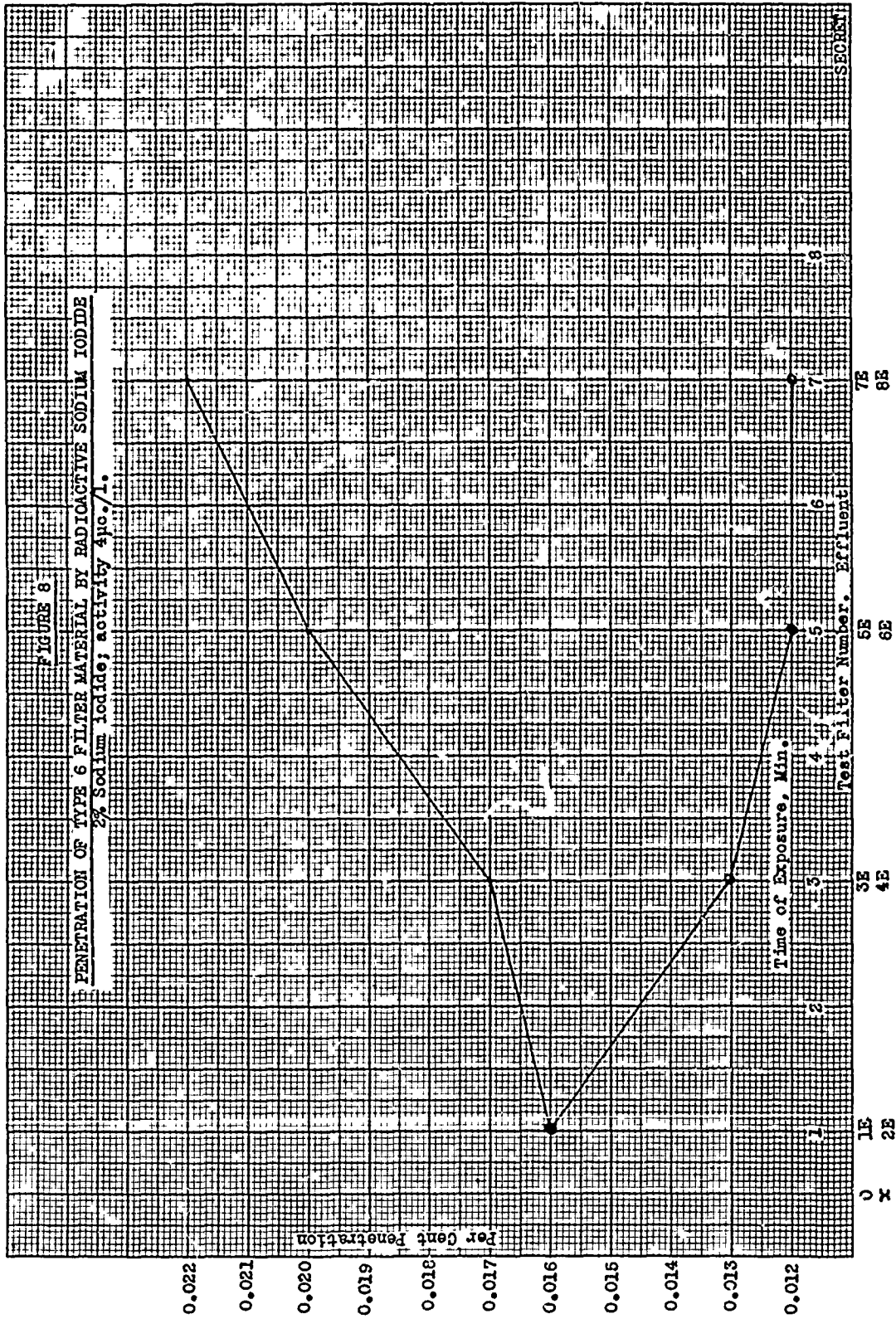
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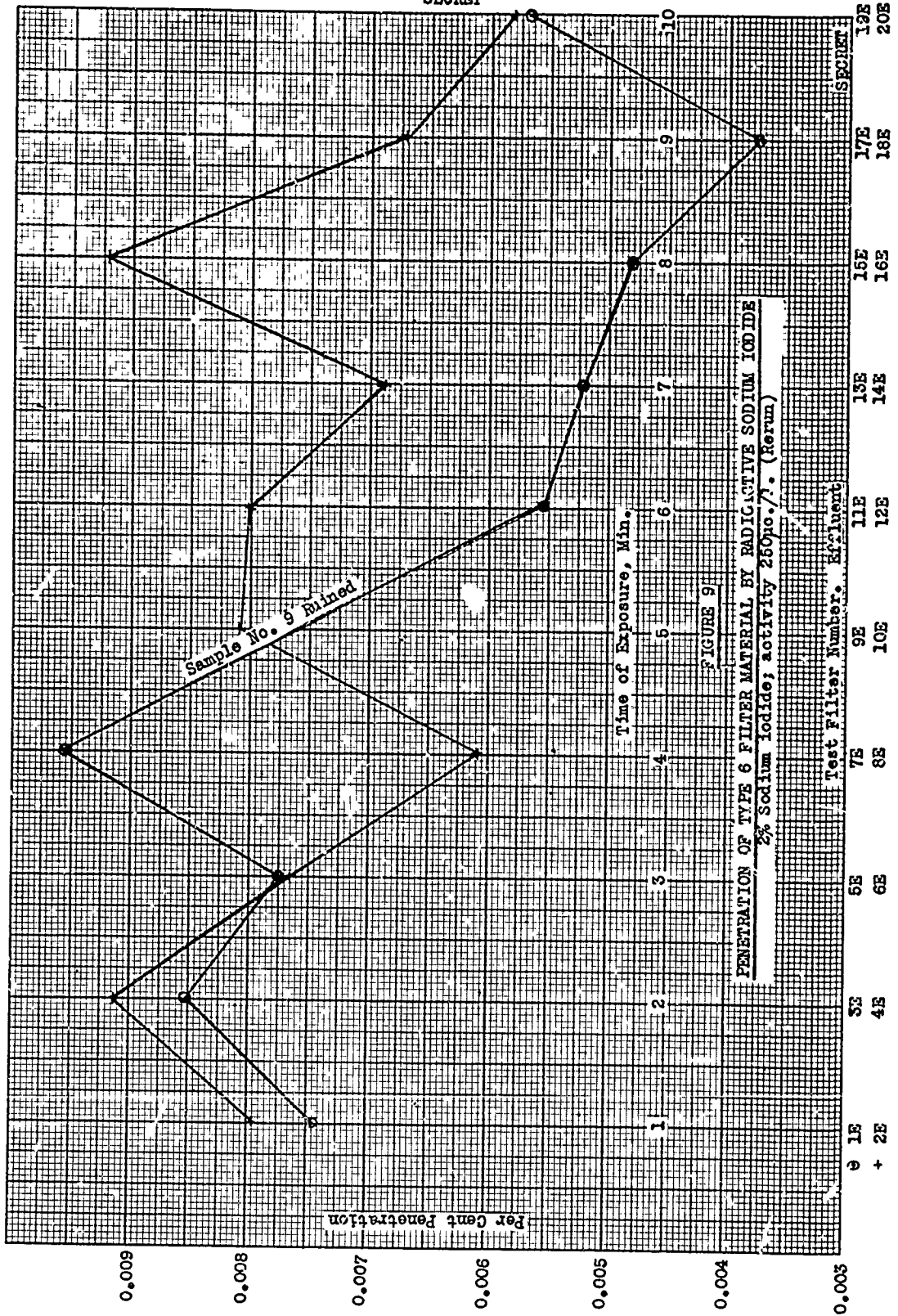


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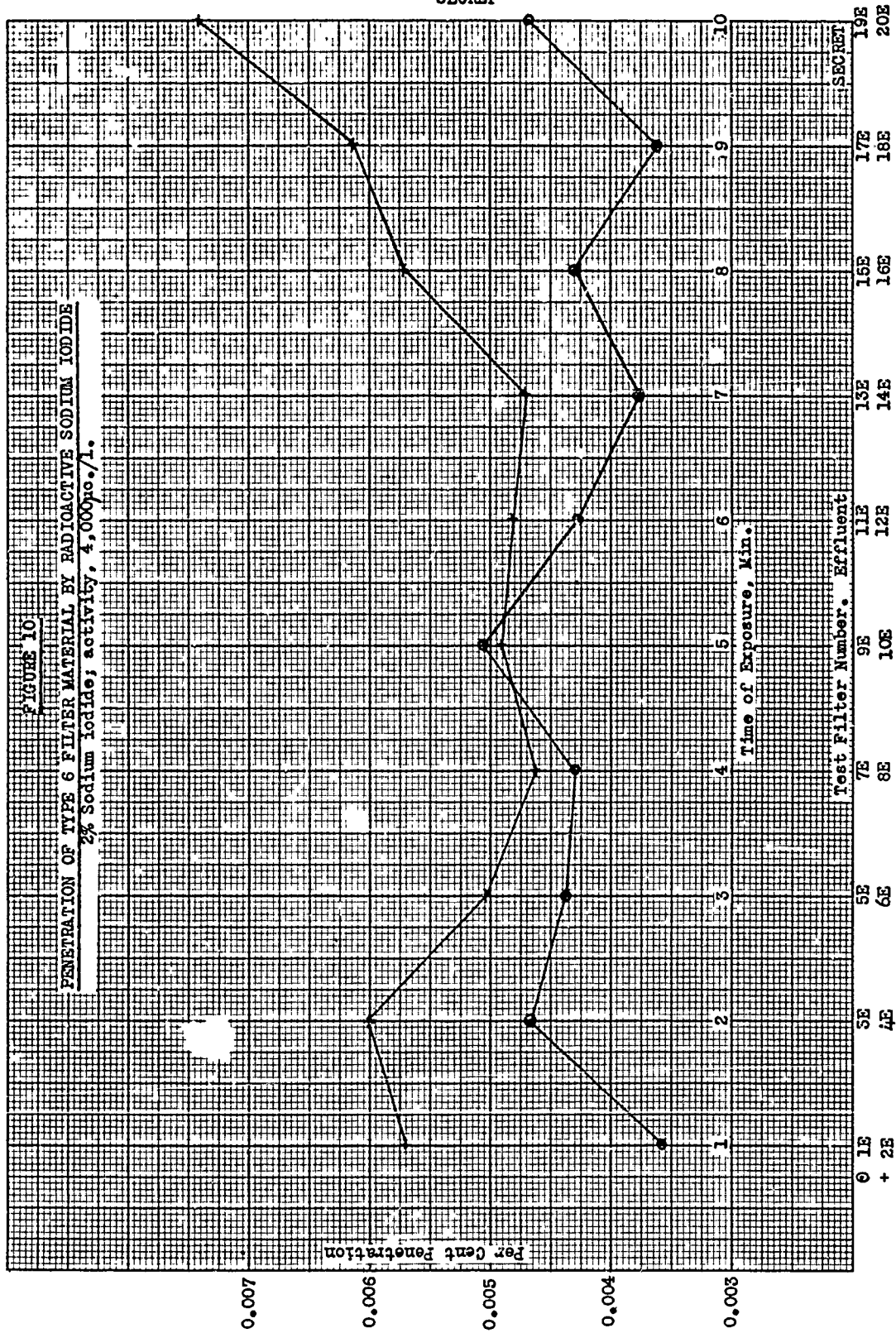


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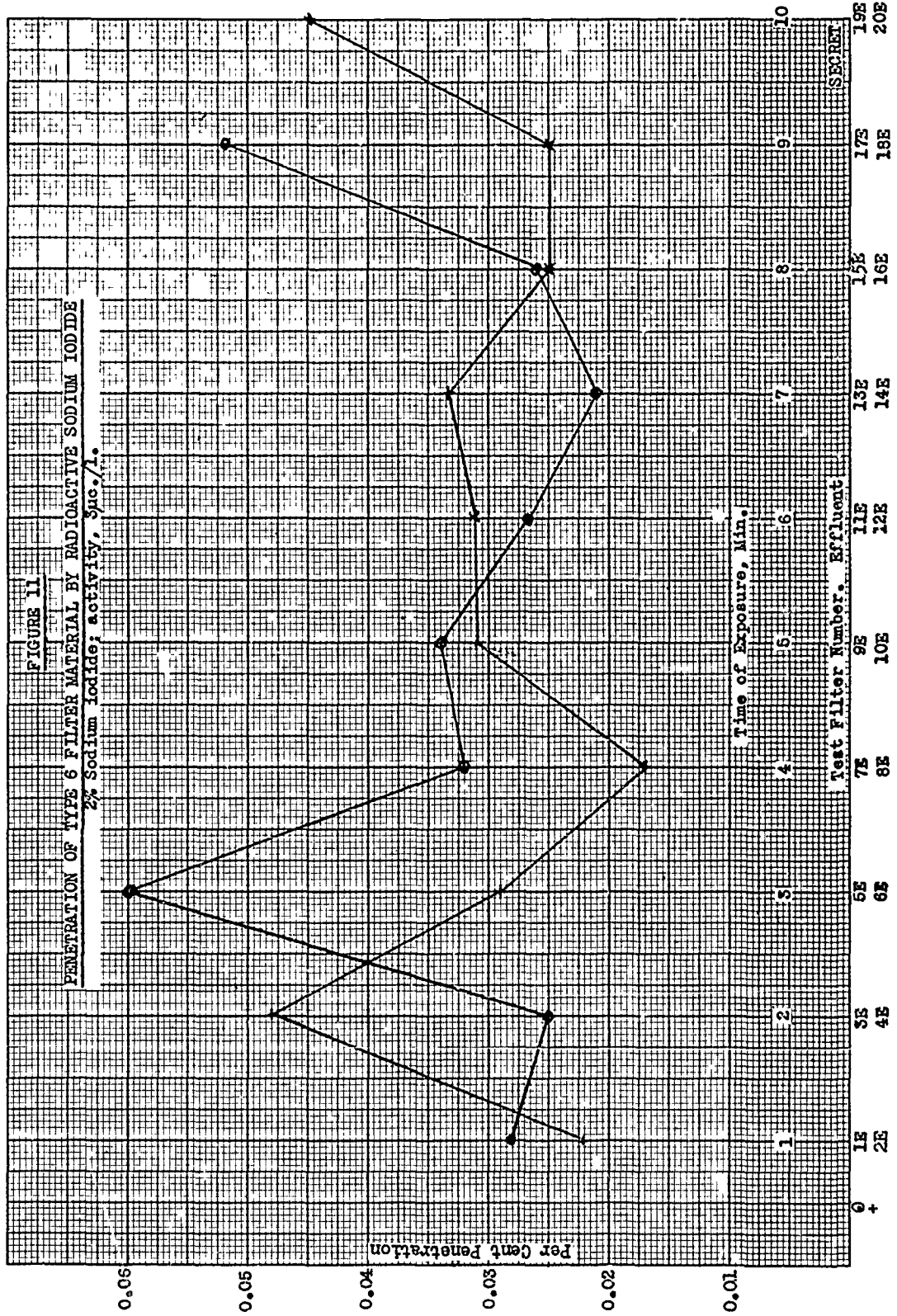
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this filter were the same, within experimental error, as those of the filter samples in holder No. 3. The filters in holder No. 3 were changed each minute of exposure and used as an influent test filter to count, whereas the filter in holder No. 4 was not changed during an entire run.

V. DISCUSSION.

The over-all accuracy of the results with 2% solution is $\pm 20\%$, although data in individual runs were much more consistent.

The exact aerosol radioactivity at which filter penetration changed was not determined, although it appears to be at a level less than $60\mu\text{c./l.}$ The reproducibility of results on changing back and forth from high to low levels of radioactivity was considered a more critical question. This reproducibility permits confidence in the results.

Possible timing errors in exposing filter samples and possible high-count errors of previous work were practically nil in the improved equipment and counting method.

VI. CONCLUSIONS.

1. The Chemical Corps type 6 filter material may be effectively used against airborne radioactive particulates of a radioactivity level of the order of magnitude of the activity anticipated in a cloud in the vicinity of a nominal atomic detonation at $H + 10$ min.

a. The penetration of type 6 filter material by an aerosol with a 0.6 Mev beta radiation and a 0.37 Mev gamma radiation in activity levels up to $20\mu\text{c./l.}$ is approximately 0.02% to 0.04%.

b. Between $60\mu\text{c./l.}$ and $4,000\mu\text{c./l.}$ aerosol activity, the penetration efficiency is approximately 0.005% to 0.007%.

2. There is a significant increase in efficiency of type 6 filter material for the removal of an aerosol in activity levels of $60\mu\text{c./l.}$ and $4,000\mu\text{c./l.}$

VII. RECOMMENDATIONS.

None, since work is continuing under this project.

VIII. BIBLIOGRAPHY.

1. Mr. E.H. Engquist and Dr. J.J. Mahoney, TCR 60, Interim Report on the Development of the Radioactive Sodium Iodide Test for Filter Material.

2. Summary Technical Report of NDRC, Div. 10, Vol. I, Aerosols and Real Gases, OSRD, pp. 640-643.

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APPENDIX A

LOSS OF ISOTOPE ON CONCENTRATION OF
ATOMIZING SOLUTION BY HEAT

Several concentrations and tests were made to determine if water could be distilled off the atomizing solution without loss of isotope. One such experiment is described.

STEP 1

Twenty-five milliliters of atomizer solution was made up containing about 5 mc. of NaI^{131} labeled in 2% inert NaI .

STEP 2

Two-minute exposures for influent samples 13, 14, 15, and 16 were made in the usual way described in experimental procedure.

STEP 3

To within 0.1 ml., the volume left after step 2 was measured to be 24.5 ml.

STEP 4

Twenty-five milliliters of distilled water was added and then the atomized liquid was heated by the infrared heat lamps until 21.5 ml. was left in the atomizer bottle. Efforts were made to stop at the same concentration at which step 3 ended, i.e., 24.5 ml. However, stopping the boiling at a volume of 21.5 ml. would have no effect on the question of isotope being lost or not by boiling.

STEP 5

Four more test filters, nos. 17, 18, 19, and 20 were made in exactly the same way as the original ones of step 2.

STEP 6

The ratios of the counts/sec. for the filters before the distillation of step 2, and after the distillation of step 5, are compared in the data.

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Before Boiling - Steps 2 and 3

Sample	Counts	Seconds	Shelf	Sample	Counts	Seconds	Shelf
13	2,048	38.4	5	14	2,048	35.5	5
13	2,048	40.0	5	14	2,048	36.8	5
13	2,048	38.9	5	14	2,048	35.4	5
13	2,048	41.0	5	14	2,048	36.4	5
15	2,048	33.5	5	16	2,048	37.2	5
15	2,048	33.2	5	16	2,048	38.0	5
15	2,048	33.4	5	16	2,048	38.2	5
15	2,048	33.2	5	16	2,048	39.4	5

Atomizer volume at start, 25 ml.

Atomizer volume at finish, 24.5 ml.

Average time for 2,048 counts was 36.8 sec.

Average count/sec. was 55.5

Appendix A

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After Boiling - Step 4

Sample	Counts	Seconds	Shelf	Sample	Counts	Seconds	Shelf
17	2,048	34.8	5	18	2,048	31.0	5
17	2,048	33.4	5	18	2,048	30.0	5
17	2,048	34.0	5	18	2,048	29.4	5
17	2,048	34.8	5	18	2,048	29.8	5
19	2,048	35.8	5	20	2,048	36.4	5
19	2,048	37.2	5	20	2,048	36.2	5
19	2,048	35.8	5	20	2,048	35.8	5
19	2,048	36.2	5	20	2,048	35.8	5

Atomizer volume at start, 21.5 ml.

Average time for 2,048 counts was 34.1 sec. Average counts/sec. = 60.0
or $60.0 \times \frac{21.5 \text{ ml.}}{(25.0 - 0.5) \text{ ml.}} = 52.7 \text{ counts/sec. for 25 ml.}$

Conclusion: There was no significant loss of isotope due to the distillation.

This conclusion is justified by comparing the 55.5 counts/sec. to 52.5 counts/sec. for before and after boiling, respectively.

Background count of 0.4 counts/sec. is negligible in the ratio of 55.5 to 52.7 counts/sec.

The probable statistical counting error for 2,048 counts is 1.3%

Appendix A

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APPENDIX 3

SAMPLE DETERMINATION OF COUNTING RATIO DUE TO
DISSOLVING NaI ON SAMPLE FILTERS IN WATER

Two influent filter samples 1I and 2I were collected simultaneously by the regular method.

Filter 1I was put in the regular aluminum cup and counted.

Filter 2I was placed in 50 ml. of distilled water. After the NaI was dissolved, 1 ml. of this solution was pipetted off and prepared for counting as described previously.

Sample number	Total counts	Total time	Counts/sec.	Counts/sec. corrected for back-ground	Counts/sec. corrected for resolving time	Ratio
1I	12,288	37.6	326.8	327	350	38.6
2I	1,536	157.8	9.73	9.08	9.08	

The above ratio is equal to:

$$\frac{\text{counts/sec. of dry filter in closed aluminum cup}}{\text{counts/sec. of residue from 1 ml. of solution in glass cup}} = \frac{350}{9.08} = 38.6$$

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APPENDIX C

CALIBRATION DATA FOR COUNTING APPARATUS

An I^{131} sample on filter paper was prepared and counts/sec. were determined on all five shelves.

Shelf no.	Total counts	Time	Counts/sec.	Counts/sec. corrected for resolving time of 200 μ c.	Counts/sec. corrected for background
Background	86.2	48.0	0.56	0.56	-
1	12,288	42.6	288	306	306
2	6,144	46.4	132	136	136
3	6,144	89.4	68.8	68.8	68.2
4	3,072	77.2	39.8	39.8	39.2
5	3,072	123.2	24.9	24.9	24.3

A comparison of counts on all shelves using shelf no. 5 as a reference is shown:

<u>Shelf</u>	<u>Relative counts/sec.</u>
1	12.6
2	5.58
3	2.81
4	1.61
5	1.00

The procedure is the same as that described in TCR 60.

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Filter material, type 6
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REPLY TO
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ERRATA

29 April 1994

MEMORANDUM FOR The Defense Technical Information Center (DTIC)
Cameron Station, Alexandria, VA 22304-6145

SUBJECT: Reports Approved for Public Release

1. The following reports have been approved for release to the public:

a. CRLR 14, Project 4-12-75-001, Filter Material Efficiency Against High Levels of Airborne Radioactive Contamination, AD-473289.

b. CRLR 148, Project 4-80-12-004, Final Engineering Test No. 51, Filter, Gas and Particulate, 1200 CFM, E26, ~~AD-473289~~.

2. Please direct any questions to the undersigned on DSN 584-2914, or (410) 671-2914.

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Sandra J. Johnson
SANDRA J. JOHNSON

Chief, Technical Releases Office

151, 20/14